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Solid-State Structures and Properties of Yttrium Hydride

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Abstract—The structural, electronic and thermodynamic properties of rare-earth hydride metal YH_2 with the cubic structure have been investigated by using the First-principles calculations within the both local-density and generalized gradient approximations based on the density-functional theory (DFT). In this study we would like discussed the bonding properties and enthalpy of formation of this hydride on the basis of electronic total and partial densities of states plots using ABINIT code.

Keywords—hydrogen storage, rare earth hydride, Yttrium hydride, density functional theory.

I. INTRODUCTION

Hydrogen appears more and more to be an attractive alternative energy carrier with hydride storage as a key element in systems approaches of renewable energies. Industrial participation is beginning to emerge and shows good potential for expansion. As members of metal hydrides, the rare-earth hydrides have received tremendous attention due to their potential application for hydrogen storage technology [1, 2]. The metals hydrides such as yttrium, cerium and erbium are promising compound in the technology of hydrogen. This great interest makes them attractive objects for studying hydrogen behavior in metal-hydrogen composition. The criteria of hydride formation are described by many researchers [3-6].

Several works are done in the rare earth hydride metal field; Griessen et al. [7] discovered that yttrium-, lanthanum- and rare earth-hydride films exhibit remarkable optical properties near their metal-insulator transition: the dihydrides are metallic and shiny while the trihydrides are semiconducting and transparent. Schoenes et al. [8] have reviewed quantitative IR and Raman studies of yttrium-hydride films and they conclude that IR data indicate that YH_3 is a substantially ionic compound. Stepanov et al [9] reported on the synthesis of Y nanoclusters and a particular chemical solid-state reaction of these clusters and used a novel type of laser ablation cluster source to synthesize Y-clusters of about 25–30nm average size at different Ar background gas pressures to hydrogenize them in situ. Garces et al. [10] investigated theoretically the ordering of hydrogen in yttrium focusing their research on the

relation between chain ordering of H, the relaxation of internal coordinates, and the electronic properties for Yttrium hydride. For each concentration, they relaxed the internal positions of all atoms in the cell and studied the density of states and the Fermi surface. First-principles electronic structure calculations have been performed for YH_2 by Jäger et al. [11] and the bonding properties of this compound were discussed on the basis of local partial densities of states and electron density plots.

The aim of this paper is to contribute to these investigations using ABINIT code based on pseudo-potentials and plane waves. In the next section we describe models and calculation method. The details of our investigation are presented in section III. The main results are summarized in the conclusion.

II. METHOD AND COMPUTATIONAL DETAILS

The electronic structure calculations are based on the density-functional theory (DFT) [12] using the local-density approximation (LDA) of Troullier and Martins [13] to approximate the exchange-correlation energy and generalized gradient approximation (GGA) have been performed by ABINIT code based on pseudo-potentials and plane waves. The electronic wave functions were expanded in plane waves up to a kinetic energy cutoff of 50 Hartree. Integrals over the Brillouin zone were approximated by sums of $8 \times 8 \times 8$ mesh of special k-point.

III. RESULTS AND DISCUSSIONS

A. Structural features

The crystal structure of YH_2 is known and occurs in a face centered cubic (fcc) with a space group of Fm-3m (225) (see figure 1). In the table 1 we summarized the relaxed structural parameters and compared them to the experimental work.



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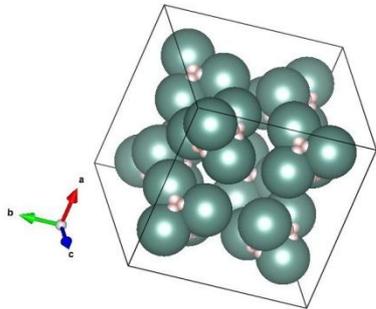


Fig. 1: Crystal structures of YH₂, big, and small circles denote Y and H atoms respectively

The deviations between the experimental and the calculated unit-cell parameter *a*, are estimated by 2.02% using GGA and 5.55% using LDA. These deviations are acceptable and are typical for the state-of-art approximation of the density functional theory. It is also noted that in this case, both LDA and GGA overestimate the lattice parameters, but GGA is more accurate than LDA. These results show that the method used in this study is reliable thus the optimized lattice constants will be used to calculate other properties.

TABLE I

RELAXED STRUCTURAL PARAMETERS AND COMPARED WITH EXPERIMENTAL WORK

	Our work		Experimental
	LDA	GGA	
<i>a</i> (Å), YH ₂ (225, Fm-3m)	5.4410	5.2591	5.1548 [14]

B. Bonding distance

In Table 2, we reported the bonding distances of each element that composes the YH₂ compound and compared with ref [14]. One can note that it's in agreement with experimental result.

TABLE II

CALCULATED BONDING DISTANCE COMPARED WITH EXPERIMENTAL WORK

YH ₂	Our work		Experimental
	LDA	GGA	
Y-H (Å)	2.35602	2.27726	2.23209 [14]

C. Enthalpy of formation

For the enthalpy formation calculation of YH₂ compound, we considered the following reaction:



We subtracted the total energies of the pure elements Y (in hexagonal structure) and the hydrogen molecule from their hydride YH₂.

In table 3, we reported the total energy and the formation enthalpy computed of YH₂ for reaction (1) for LDA and GGA approximation. The total energy of the hydrogen molecule and has been also calculated. We noted that the heat formation calculated with LDA approximation is less than the one calculated with GGA and it is in good agreement with experimental results.

TABLE III
CALCULATED HEAT OF FORMATION OF YH₂

Elements	Total Energy (eV)		Enthalpy of formation (kJ/mol)	
	LDA	GGA	LDA	GGA
Y	-55.88	-43.83	-	-
H ₂	-30.82	-31.40	-	-
YH ₂	-89.05	-78.04	-226.74	-271.12
			-226.7 [15]	
Experimental			-227.5 [16]	
			-224.5 [4]	

D. Electronic density of states (TDOS and PDOS)

To investigate the electronic structure of the rare hydride compound YH₂, we used the calculated lattice parameters reported in Table 1.

Figure 2 showed the total density of states. The electronic structure is metallic without the energy gap because the DOS reach a value of 0.8306 (states/eV/cell) at the Fermi level of -5.34eV.

The partial density of states is plotted in figure 3. In the low energy section (from -7 to -12 eV in figure 2) we can see two peaks; the first concerns H-s/Y-d bonding interaction and it's followed by a second peak primarily produced by the H-s states. Y-s and Y-p have a very small contribution to the valance band from the region -12 (eV) to 0.



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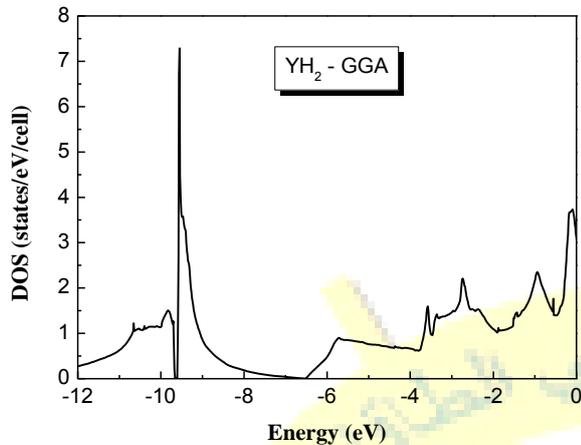


Fig. 2: The total density of state of YH₂

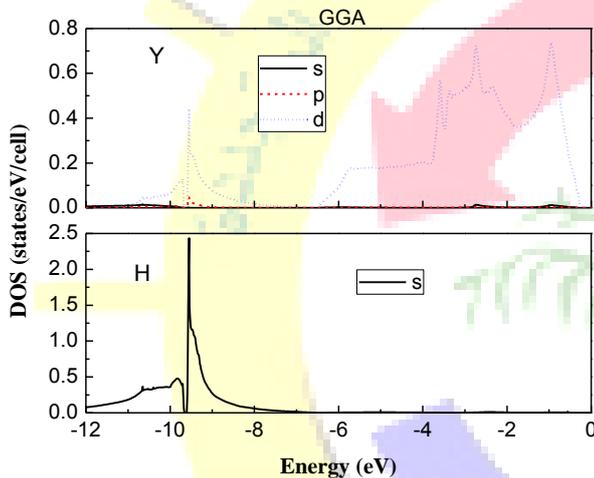


Fig. 3: The partial density of state of YH₂

IV. CONCLUSION

In this work we investigate the electronic structure calculations for rare earth hydride YH₂ using the ABINIT code. The electronic structure is obtained from GGA. The enthalpy formation for this rare earth hydride compound is calculated for two approximations, local-density approximation and generalized gradient approximation. This allowed us to better analyze our compound by studying the total and partial density of states. Also from the plot of the DOS, we noted that YH₂ has no gap which means that our compound is a metallic and the valence band is dominated by hydrogen atoms.

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